URS 757-5

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SOME PROPERTIES OF RADIOACTIVE FALLOUT:
SURFACE DETONATION COULOMB C

Final Report

September 1969

Contract No. DAHC20-69-C-0142
OCD Work Unit 3119A

URS RESEARCH COMPANY

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URS 757-5

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Final Report

September 1969

by

Carl F. Miller
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155 Bovet Road
San Mateo, California 94402

for

OFFICE OF CIVIL DEFENSE
Office of the Secretary of the Army
Department of the Army
Washington, D.C. 20310

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OCD REVIEW NOTICE

URS 757-5
SUMMARY

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## SUMMARY

Data on the ionization-rate decay, activity-particle size distributions, and exposure rate-conversion coefficient for soil and filter-collected samples of fallout from Shot Coulomb C have been presented. Radiochemical analyses of selected samples permitted the correlation of the results in terms of equivalent fission levels relative to the Mo-99 content of the samples. The gross fractionation ratio relative to normal U-235 fission products was generally lower than observed for the fallout from larger yield explosions at all times from 240 to 6,000 hours after detonation. Individual radionuclide radiochemical "R" values were determined for Sr-89, Zr-95, Te-132, and Ba-140. No soil induced activities were detected to be present in significant amount at the time the analyses were carried out at around H + 270.

# INTRODUCTION

The surface detonation named Coulomb C was part of the PLUMBBOB weapons test series conducted at the Nevada Test Site in the summer of 1957; this surface detonation has been assigned a detonation yield of 0.5 KT.[1] In this report, data on the decay of the fallout on activity-particle size distributions, on radiochemical analyses, and on the ionization rate conversion factor are reported.

No fallout collection and analysis program was sponsored for this event. The author and Mr. George T. Anton were visiting the test site for another purpose at the time and upon learning of the fallout pattern from this detonation, obtained permission from the site director to enter the area along with Mr. William J. Brady of the Reynolds Electric Company Radiological Safety Office for the purpose of taking some surface soil samples. In addition, a vacuum filter unit was used to pick up particles from the surface of a macadam road which crossed the fallout pattern. The samples were sent to the U.S. Naval Radiological Defense Laboratory, San Francisco, California, for radiochemical analyses, gross assay of radioactive content, sieve analysis, and measurement of the decay rates.

## RADIOCHEMICAL AND ASSAY RESULTS

Six soil samples were taken, each covering a square foot in area and about 1-inch in depth; three filter samples were taken at three points along the road crossing the pattern. The first two soil samples were taken on the hotline of the fallout pattern at a distance of 0.4 miles from ground zero where the exposure rate at 3 feet above the surface was measured to be 4 R/hr at 48 hours after detonation. The second set of two soil samples were taken on the hotline at a distance of 0.5 miles from ground zero where the exposure rate was 3.5 R/hr at 48 hours after detonation. And the third set of two soil samples were taken on the hotline at a distance of 1.8 miles from ground zero where the exposure rate was 660 mR/hr at 48 hours after detonation. The distance from ground zero to the road from which the vacuum filter samples were obtained was about two miles.

The gross radioactive content of all the samples was measured in the USNRDL Doghouse counter[2,3] and then two of the soil samples were aliquoted for radiochemical analysis and decay measurements. Only one of the filter-collected samples contained sufficient activity for similar analyses. One of the soil samples was used to measure the decay rate in the Doghouse counter. The remainder were subjected to sieve analysis. The sample designations, relative gross activity corrected to 240 hours after detonation, and disposition of each sample are summarized in Table 1.* The results of the radiochemical analyses of the aliquoted samples are given in Table 2. The $i_d$ and $i_g$ values for the aliquots indicate that some difficulties occurred in obtaining good separation of the Mo-99 from the soil samples (probably due to the bulk of foreign substances present in the solutions). The C9 sample analysis was done in triplicate and had much less inactive soil present; the $i_g$ value from this sample was therefore used to derive $i_g$ values for the decay samples and as a basis for estimating the $a_f$ values for all the samples.

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* The analyses were carried out by D. Love, D. Sam, P. Strom, M. Nucholls, and D. MacDonald in December, 1957 and were reported in a USNRDL memorandum to the author by Paul Zigman on January 21, 1958.

Table 1

DESIGNATION, GROSS ACTIVITY, AND DISPOSITION

OF COULOMB C SAMPLES

| Sample Designation | Type of Sample | $a_d$ (cpm at H + 240) | Total Weight (gm) | Disposition |
|---|---|---|---|---|
| C1 | Soil | 661,500 | 1,366.5 | 110.8 gm for analyses; remainder sieve analyzed |
| C2 | Soil | 933,200 | - | not treated, retained for future analyses as needed |
| C3 | Soil | 808,700 | - | sieve analyzed |
| C4 | Soil | 801,900 | 1,097.8 | 62.1 gm for analyses; remainder sieve analyzed |
| C5 | Soil | 161,700 | - | sieve analyzed |
| C6 | Soil | 143,700 | - | Doghouse decay |
| C7 | Filter | 1,0[illegible]7 | - | not treated |
| C8 | Filter | 3,283 | - | not treated |
| C9 | Filter | 55,030 | - | entire sample subjected to radiochemical and other analyses |

Table 2

SUMMARY OF RADIOCHEMICAL ANALYSIS AND ASSAY RESULTS ON COULOMB C FALLOUT SAMPLE ALIQUOTS

| Quantity Measured | Aliquot Designation | | | | | |
|---|---|---|---|---|---|---|
| | C1a | C1b | C1c[a] | C4a | C4b[a] | C9[a] |
| Weight (gm) | 37.84 | 32.05 | 40.90 | 30.79 | 31.30 | - |
| $a_d$ (cpm at H + 240) | 12,590 | 13,730 | 16,500 | 34,080 | 15,380 | 55,030 |
| $a_g$ ($10^{-9}$ ma at H + 240) | 123 | 134 | 165 | 345 | 150 | 511[b] |
| $a_g/a_d$ ($10^{-12}$ ma/cpm) | 9.77 | 9.76 | 10.00 | 10.12 | 9.75 | 9.29 |
| $a_f$ ($10^{13}$ fissions)[c,d] | 6.28 | 5.15 | - | 12.70 | - | 15.58[e] |
| $i_d$ ($10^{-9}$ cpm/f at H + 240) | 0.200 | 0.267 | - | 0.268 | - | 0.353 |
| $i_g$ ($10^{-20}$ ma/f at H + 240) | 0.196 | 0.260 | - | 0.272 | - | 0.328[e] |

a Aliquots C1c, C4a, and 1/10 of the dissolved portion of sample C9 were used for decay measurements in the ion chamber.

b $a_g$ for the dissolved portion of the sample at H + 268 was $323 \times 10^{-9}$ ma indicating that only about 0.708 of the activity was dissolved and retained in solution (a certain loss of volatile activities including the iodine isotopes 131 and 132 would be expected at this time after detonation); $a_g(268)$ for the original sample would have been $456 \times 10^{-9}$ ma assuming a decay rate as for C4b.

c Based on Mo-99 analysis.

d Radiochemical "R" values reported were: 0.040 for Sr-89; 0.059 for Te-132; 0.23 for Ba-140 in Sample C9; and 0.51 for Zr-95 in sample C4a.

e Analysis of solution in triplicate gave $1.2 \times 10^{14}$ fissions for the portion dissolved; this would give an $i_g$ value of $0.426 \times 10^{-20}$ ma/fission at H + 240 which is too high. Assuming partial solution of the sample, $a_f$ would be given by $(456/323) \times 1.2 \times 10^{4}$ or $1.69 \times 10^{14}$ fissions. Using $0.328 \times 10^{-20}$ ma/fission at H + 240 as derived for aliquot C4b in Table 3 gives an $a_f$ value of $1.56 \times 10^{14}$ fissions; the latter value was selected since it is apparent that at least some activities were lost in the dissolution process.

The $a_f$ value for the decay sample C9a, at 0.1 volume of the solution from C9 is $1.20 \times 10^{13}$ fissions. Comparing the decay curve measurements in the time period of 2,000 to 6,000 hours after detonation for samples Clc and C9a gives a ratio of $a_f$(Clc) to $a_f$(C9a) of 4.14 and, in the time period of 400 to 1,300 hours after detonation for samples C4b and Clc, a ratio of $a_f$(C4b) to $a_f$(Clc) of 0.920. The estimated $a_f$ values from these ratios are therefore $4.97 \times 10^{13}$ fissions for sample Clc and $4.57 \times 10^{13}$ fissions for sample C4b; the $i_g$ and $r_{fp}$ values for the three samples at different times after detonation are given in Table 3. The data for aliquot Clc is plotted with time after detonation in Figure 1; the curve has the characteristic curvature to a steeper slope starting at 2,500 hours after detonation. The slope of the straight line portion of the curve from about 300 to 2,000 hours after detonation is -0.86.

The variation of the gross fractionation ratio, $r_{fp}$, with time after detonation for each of the three aliquots is shown in Figure 2. The shape of the curves are similar to those given in Reference 2 for the fallout from the tower detonations Diablo and Shasta. The $r_{fp}$ values for the dissolved portion of sample C9 are lower than those for the two soil samples until about 2,000 hours after detonation due to apparent losses during the dissolution process of such nuclides as I-131, I-132, Ru-103, Rh-105, and perhaps Te-132 all of which would be present in an unfractionated FP mixture in significant quantity at H + 240 to H + 270. At this time, about 27 percent of the gamma ray energy would be contributed by the Iodine isotopes and the Lanthanum isotopes would contribute about 51 percent at H + 240. Up to about 1,000 hours after detonation the effect on $r_{fp}$ of losses in both the I and Ru(Rh) isotopes would be possible; after about 1,500 hours after detonation, effects on the value of $r_{fp}$ could only be due to Ru(Rh) isotopes and these could persist at times longer than 6,000 hours after detonation (i.e., at times beyond the time at which the Zr(Nb) isotopes are the predominating contributors to $i_g$). The differences between the $r_{fp}$ values for aliquot Clc and C4b are very small in the period of 240 to 1,270 hours after detonation.

With the decay data of Table 3, revised values of $a_f$ and $i_d$ for the various aliquots are as follows:

Table 3

SUMMARY OF IONIZATION-RATE DECAY MEASUREMENTS AND $r_{fp}$ VALUES OF COULOMB C FALLOUT SAMPLES[a]

| Time After Detonation (hours) | $i_{\gamma}$ ($10^{-20}$ ma/fission) Sample Aliquot Clc | C4b | C9a | Reference for Unfractionated U-235 FP | $r_{fp}$ Sample Aliquot Clc | C4b | C9a |
|---|---|---|---|---|---|---|---|
| 240 | 0.332 | 0.328 | | 1.00 | 0.332 | 0.328 | |
| 242 | 0.330 | | | 0.995 | 0.332 | | |
| 243 | | 0.324 | | 0.985 | | 0.329 | |
| 245 | 0.326 | | | 0.960 | 0.340 | | |
| 260 | 0.310 | 0.304 | | 0.915 | 0.339 | 0.332 | |
| 268 | 0.300 | 0.293 | 0.269 | 0.885 | 0.339 | 0.331 | 0.304 |
| 290 | 0.278 | 0.271 | | 0.808 | 0.344 | 0.335 | |
| 319 | 0.252 | 0.247 | | 0.724 | 0.348 | 0.341 | |
| 331 | 0.242 | 0.241 | | 0.700 | 0.346 | 0.344 | |
| 334 | | | 0.239 | 0.690 | | | 0.346 |
| 356 | 0.229 | 0.225 | 0.227 | 0.642 | 0.357 | 0.350 | 0.354 |
| 411 | 0.205 | 0.204 | 0.198 | 0.550 | 0.373 | 0.371 | 0.360 |
| 506 | 0.172 | 0.170 | 0.163 | 0.440 | 0.391 | 0.386 | 0.370 |
| 525 | 0.166 | 0.165 | 0.158 | 0.422 | 0.393 | 0.391 | 0.374 |
| 598 | 0.152 | 0.153 | 0.144 | 0.376 | 0.404 | 0.407 | 0.383 |
| 602 | 0.148 | 0.148 | 0.142 | 0.358 | 0.413 | 0.413 | 0.397 |
| 672 | 0.134 | 0.134 | 0.128 | 0.314 | 0.427 | 0.427 | 0.408 |
| 722 | 0.125 | 0.126 | 0.119 | 0.285 | 0.439 | 0.442 | 0.418 |
| 770 | 0.119 | 0.120 | 0.110 | 0.260 | 0.458 | 0.462 | 0.423 |
| 843 | 0.110 | 0.111 | 0.101 | 0.229 | 0.480 | 0.485 | 0.441 |
| 908 | 0.103 | 0.103 | (1)950 | 0.206 | 0.500 | 0.500 | 0.461 |
| 937 | (1)998 | 0.100 | (1)933 | 0.197 | 0.507 | 0.508 | 0.474 |
| 1,008 | (1)938 | (1)943 | (1)883 | 0.180 | 0.521 | 0.524 | 0.491 |
| 1,080 | (1)884 | (1)880 | (1)842 | 0.162 | 0.546 | 0.543 | 0.520 |
| 1,270 | (1)757 | (1)757 | (1)749 | 0.127 | 0.596 | 0.596 | 0.590 |
| 1,610 | (1)622 | | (1)608 | (1)888 | 0.700 | | 0.685 |
| 2,020 | | | (1)487 | (1)659 | | | 0.739 |
| 2,050 | (1)515 | | | (1)640 | 0.805 | | |
| 2,420 | | | (1)429 | (1)528 | | | 0.812 |
| 2,460 | (1)433 | | | (1)518 | 0.836 | | |
| 3,190 | (1)316 | | (1)319 | (1)375 | 0.843 | | 0.851 |
| 3,910 | (1)246 | | | (1)286 | 0.860 | | |
| 4,620 | (1)185 | | (1)188 | (1)211 | 0.877 | | 0.891 |
| 4,800 | | | (1)173 | (1)195 | | | 0.887 |
| 4,920 | (1)164 | | (1)162 | (1)185 | 0.886 | | 0.876 |
| 5,640 | (1)121 | | (1)121 | (1)139 | 0.870 | | 0.870 |
| 6,410 | (2)884 | | (2)858 | (1)104 | 0.850 | | 0.825 |
| 7,100 | (2)654 | | (2)628 | (2)780 | 0.838 | | 0.805 |
| 7,920 | (2)468 | | (2)425 | (2)570 | 0.821 | | 0.746 |
| 8,660 | (2)335 | | | (2)430 | 0.779 | | |
| 9,450 | (2)242 | | | (2)325 | 0.745 | | |
| 10,120 | (2)185 | | | (2)262 | 0.706 | | |
| 11,080 | (2)129 | | | (1)192 | 0.672 | | |
| 12,280 | (3)872 | | | (1)135 | 0.646 | | |

a Numbers in parentheses are the number of zeros between the decimal point and the first digit.

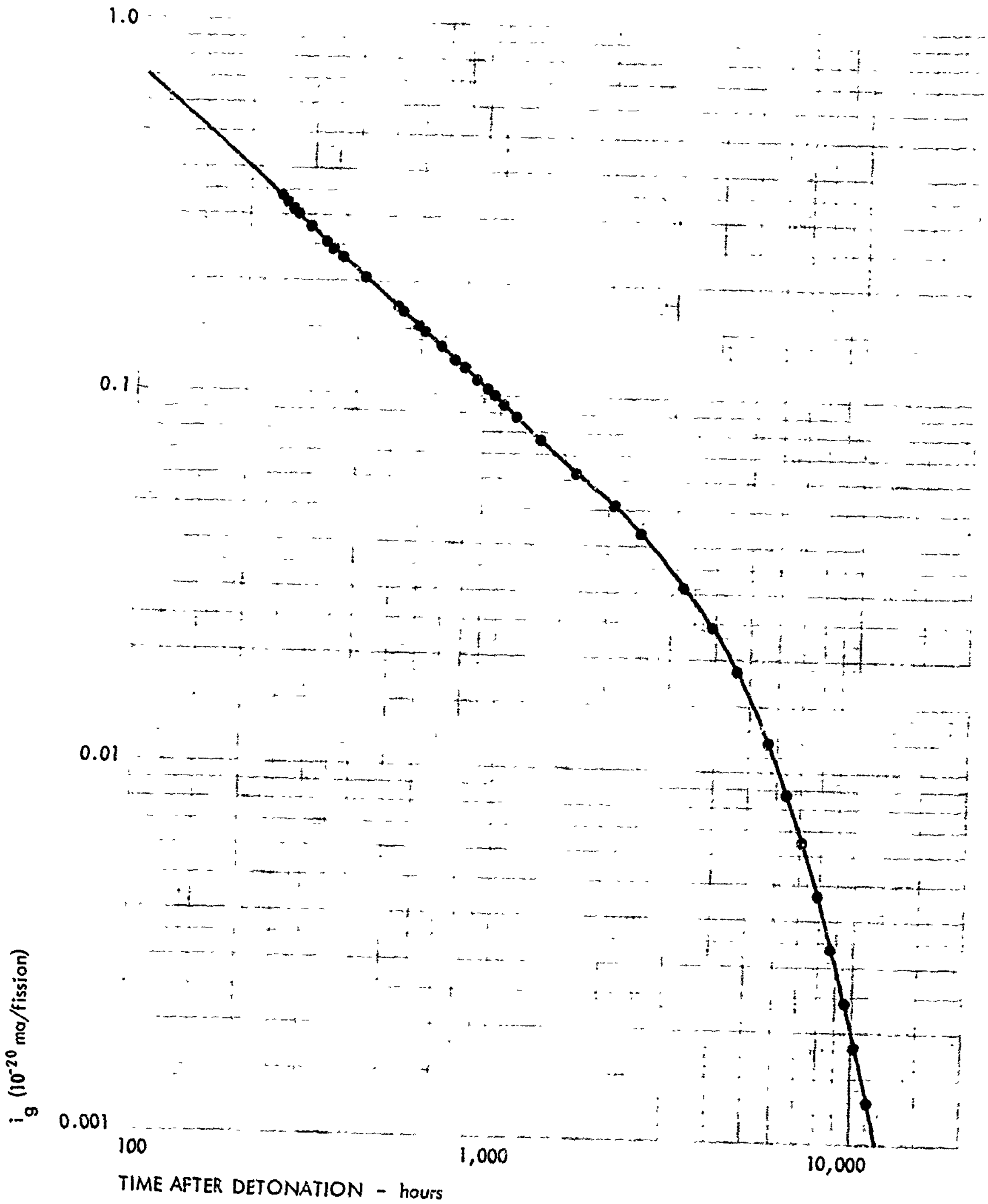


Figure 1. Ionization-Rate Decay of Aliquot Clc of Coulomb C Fallout Sample Cl.

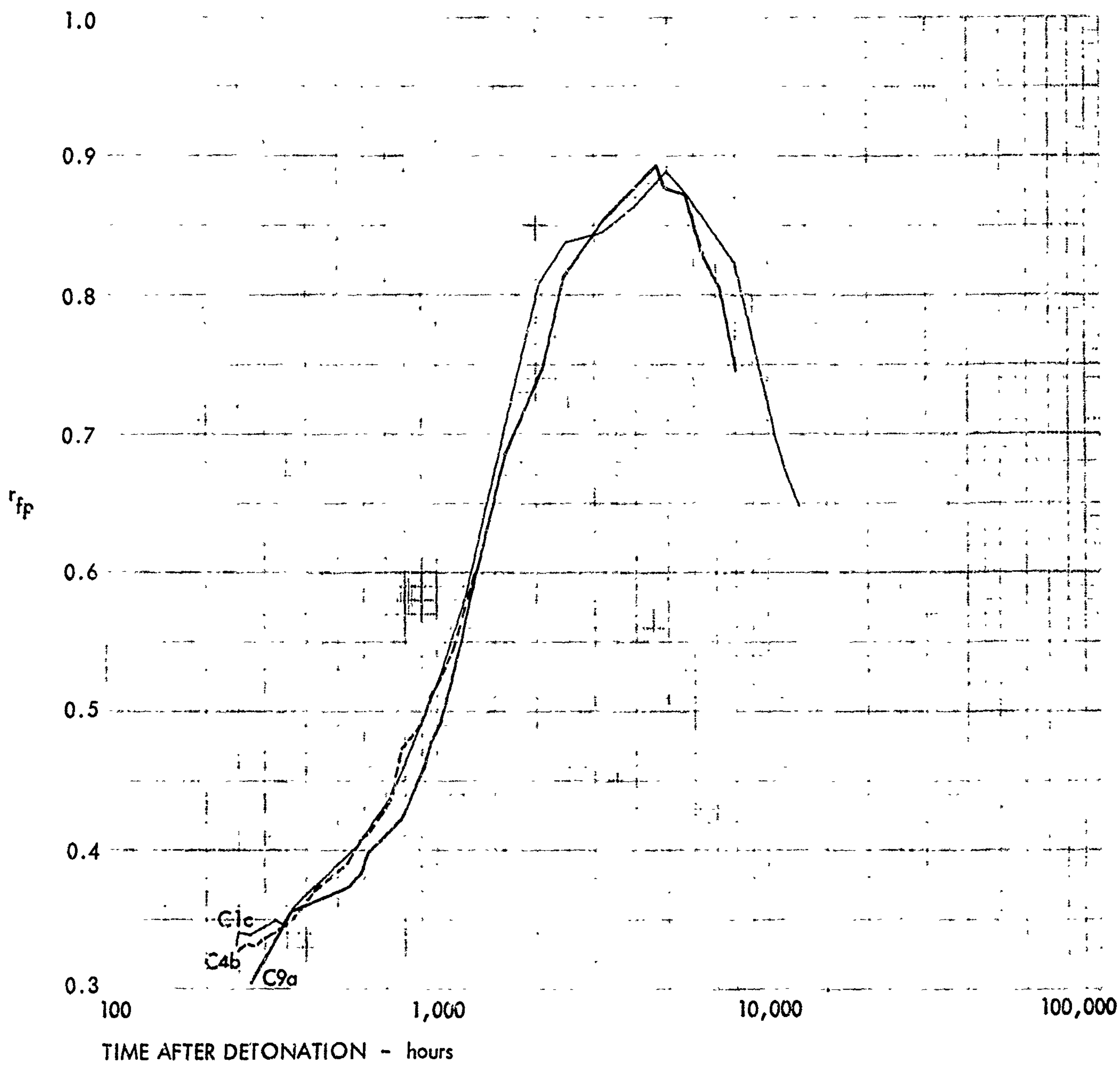


Figure 2. Variation of $r_{fp}$ with Time after Detonation for Aliquots of Coulomb C Fallout Samples.

| Aliquot | $a_f(10^{13}f)$ | $i_d(10^{-9}$ cpm at H + 240/f) |
|---|---|---|
| C1a | 3.705 | 0.340 |
| C1b | 4.036 | 0.340 |
| C1c | 4.970 | 0.332 |
| C4a | 10.52 | 0.324 |
| C4b | 4.573 | 0.336 |
| C9 | 15.58 | 0.353 |
| Average | - | 0.337 |

The Doghouse count-rate decay of Sample C6 is given along with the ratio of $i_g$ to $i_d$ in Table 4; the increase of this ratio with time from about 240 to 800 hours after detonation shows that the count-rate decay is more rapid in this time period than is the ionization-rate decay.

The average H + 240 value of $i_d$ given above was used to calculate the equivalent fission contents and surface density of activity for the three soil sampling locations using the Doghouse count-rate data given in Table 1. The results and calculated values of $K_s$, the H + 1 exposure rate activity conversion coefficient, are given in Table 5; the average value of $K_s$ for the last five samples (some of sample C1 was spilled in transfer) is 864 R/hr at 1 hr per KT/sq mi. The $K_s$ values, although in the proper range, may be in error by as much as 15 percent due to the errors involved in estimating the decay correction factor (not considering other sources of error).

Table 4

DOGHOUSE COUNT-RATE DECAY OF SAMPLE C6

| Time After Detonation (hours) | $i_d$ ($10^{-11}$ cpm/f) | $i_g/i_d$ ($10^{-12}$ ma/cpm) |
|---|---|---|
| 240 | 33.7 | 9.79 |
| 244 | 32.9 | 9.88 |
| 300 | 24.9 | 10.8 |
| 320 | 21.7 | 11.5 |
| 332 | 20.4 | 11.8 |
| 357 | 18.5 | 12.3 |
| 410 | 15.4 | 13.2 |
| 434 | 14.2 | 13.7 |
| 504 | 12.0 | 14.2 |
| 600 | 10.1 | 15.0 |
| 672 | 8.92 | 15.0 |
| 770 | 7.75 | 15.5 |
| 844 | 7.14 | 15.5 |
| 900 | 6.81 | 15.1 |
| 938 | 6.57 | 15.2 |
| 1,007 | 6.10 | 15.4 |

Table 5

CALCULATED VALUES OF OBSERVED EXPOSURE RATE-ACTIVITY CONVERSION COEFFICIENTS FOR COULOMB C FALLOUT SAMPLE LOCATIONS

| Quantity | Sample Number C1 | C2 | C3 | C4 | C5 | C6 |
|---|---|---|---|---|---|---|
| r(mi)[a] | 0.4 | 0.4 | 0.5 | 0.5 | 1.8 | 1.8 |
| $A_f$($10^{15}$ fissions/sq ft) | 1.96 | 2.77 | 2.40 | 2.38 | 0.479 | 0.426 |
| I(R/hr at 48 hr) | 4.0 | 4.0 | 3.5 | 3.5 | 0.66 | 0.66 |
| $I_s$(R/hr at 1 hr)[b] | 450 | 450 | 400 | 400 | 75 | 75 |
| $K_s'$($10^{-13}$R/hr at 1 hr)/(f/sq ft) | 2.30 | 1.62 | 1.67 | 1.68 | 1.57 | 1.76 |
| $K_s$(R/hr at 1 hr)/(KT/sq mi) | 1,200 | 840 | 870 | 870 | 820 | 920 |

a r is the distance from ground zero to sampling locations on the fallout pattern hot line.

b the decay correction factor was calculated using the assumption that the ratio of the $r_{fp}$ curve for Coulomb C fallout to that for the Diablo/Shasta fallout remained the same at 1 and 48 hours after detonation as it is for 240 hours after detonation; this assumption gives $i_g(1)$ = 248$\times10^{-28}$ ma/fission and $i_g(48)$ = 2.19$\times10^{-20}$ ma/fission which results in a decay correction from 48 to 1 hour of 113. A $t^{-1.2}$ decay correction factor would be 104 from 48 to 1 hour after detonation; the $r_{fp}$ value associated with the indicated values of $I_s$ is 0.230.

ACTIVITY DISTRIBUTIONS FOR COULOMB C FALLOUT SAMPLES

Data on the sieve analysis of Samples C1, C4, and C5 were reported in memorandum form to the author by E. A. Schuert.* The samples were dry-sieved and the sieve fractions were then assayed for gross activity with the Doghouse counting system at H + 357. Many spherical particles with diameters in the range of 500 microns were observed in Sample C1. These particles appeared to be melted silicates or silica; their general color was smoke grey but a small percentage were clear glassy particles.

The count-rate data in percentage form are given in Table 6 along with the results of a distribution curve analysis as shown in Figure 3. The samples appear to consist of two distributions. The small and large particle distributions from the samples at downwind distances of 0.4 and 0.5 miles were almost the same with a fallout particle diameter range of 600 to about 4,000 microns and a median diameter of 1,600 to 1,800 microns. At the downwind distance of 1.8 miles, the diameter range for the fallout distribution is estimated to be 190 to 850 microns with a median diameter of 415. The characteristic S-shaped distribution curves are obtained when the particle diameter limits are imposed. In the case of sample C5, the background or "small particle" distribution actually contains both smaller and larger particles than the fallout or "large particle" distribution.

* Schuert, E. A., U.S. Naval Radiological Defense Laboratory memorandum to C. F. Miller, dated 30 December 1957.

Table 6

SUMMARY OF SIEVE ANALYSIS OF COULOMB C FALLOUT SAMPLES

| Diameter (microns) | Accumulated Percentage of Gamma Count Rate | | |
|---|---|---|---|
| | C1 | C4 | C5 |
| **1. Total Sample Distribution** | | | |
| > 2,000 | 100.00 | 100.00 | 100.00 |
| 840 - 2,000 | 66.96 | 59.08 | 99.97 |
| 590 - 840 | 15.06 | 13.59 | 95.43 |
| 420 - 590 | 5.54 | 4.16 | 82.31 |
| 300 - 420 | 3.84 | 2.80 | 51.11 |
| 250 - 300 | 1.45 | 1.48 | 12.19 |
| 150 - 250 | 1.45 | 1.47 | 7.64 |
| 105 - 150 | 0.35 | 0.34 | 0.94 |
| 75 - 105 | 0.16 | 0.16 | 0.49 |
| < 75 | 0.072 | 0.052 | 0.13 |
| **2. Background Distribution[a]** | | | |
| > 2,000 | 100.0 | 100.0 | 100.0 |
| 840 - 2,000 | 98.3 | 97.1 | 99.4 |
| 590 - 840 | 65.0 | 60.7 | 74.6 |
| 420 - 590 | 46.2 | 40.4 | 55.2 |
| 300 - 420 | 32.0 | 27.2 | 37.6 |
| 250 - 300 | 12.1 | 14.4 | 23.2 |
| 150 - 250 | 12.1 | 14.3 | 16.6 |
| 105 - 150 | 2.9 | 3.3 | 5.2 |
| 75 - 105 | 1.3 | 1.6 | 2.7 |
| < 75 | 0.60 | 0.50 | 0.72 |
| **3. Fallout Particle Distribution** | | | |
| > 2,000 | 100.0 | 100.0 | 100.0 |
| 840 - 2,000 | 62.7 | 54.7 | 100.0 |
| 590 - 840 | 8.25 | 8.18 | 100.0 |
| 420 - 590 | | | 88.2 |
| 300 - 420 | | | 54.1 |
| 250 - 300 | | | 9.76 |
| 150 - 250 | | | 5.67 |
| $d_{min}$ (microns) | 600 | 600 | 190 |
| $d_{50}$ (microns) | 1,600 | 1,800 | 415 |
| $d_{max}$ (microns) | ~ 4,000 | ~ 4,000 | 850 |

a About 12.0 percent of the total activity was with the background distribution for Sample C1; about 10.3 percent for Sample C4; and about 18.1 percent for Sample C5.

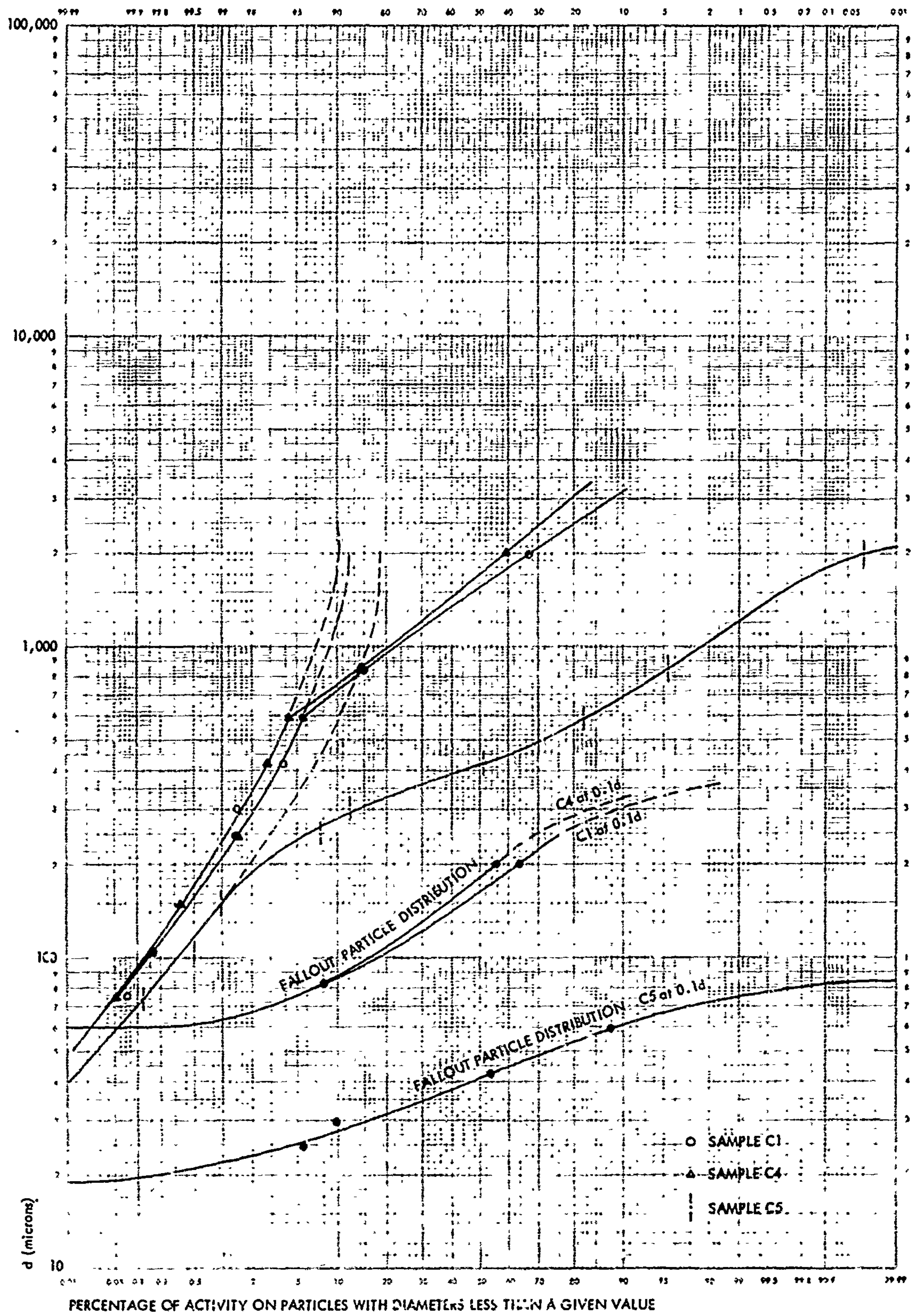


Figure 3. Activity Distribution of Soil Samples Taken from Three Different Locations on the Coulomb C Fallout Pattern.

## SUMMARY AND CONCLUSIONS

Data on the ionization-rate decay, activity-particle size distributions, and exposure rate-conversion coefficient for soil and filter-collected samples of fallout from Shot Coulomb C have been presented. Radiochemical analyses of selected samples permitted the correlation of the results in terms of equivalent fission levels relative to the Mo-99 content of the samples. The gross fractionation ratio relative to normal U-235 fission products was generally lower than observed for the fallout from larger yield explosions at all times from 240 to 6,000 hours after detonation. Individual radionuclide radiochemical "R" values were determined for Sr-89, Zr-95, Te-132, and Ba-140. No soil induced activities were detected to be present in significant amount at the time the analyses were carried out at around H + 270.

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13. ABSTRACT


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|---|---|---|---|---|---|---|
| | ROLE | WT | ROLE | WT | ROLE | WT |
| Fallout | | | | | | |
| Radioactivity | | | | | | |
| Ionization-rate | | | | | | |
| Ionization-rate decay | | | | | | |
| Exposure rate | | | | | | |
| Radiochemical | | | | | | |
| Radionuclides | | | | | | |
| Fission products | | | | | | |
| Fractionation | | | | | | |
| Exposure rate-conversion coefficient | | | | | | |
| Fallout pattern | | | | | | |
| Induced activities | | | | | | |
| Specific activity | | | | | | |

Unclassified
Security Classification